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NASA Contractor Report 156876

Nonlinear Viscoelastic Characterization of Thin Polyethylene Film

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Dr. J. S. Wilbeck

July 1981

National Aeronautics and Space Administration

Wallops Flight Center
Wallops Island, Virginia 23337
AC 804 824-3411

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Prepared Under Contract No. NAS6-3077

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- Mr. Tom Pollard for his work in obtaining the material data needed in developing and validating the material models.

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TABLE OF CONTENTS

Section	Title	Page
	SUMMARY	1
I	INTRODUCTION	3
II	VISCOELASTIC THEORY	5
	A. Viscoelasticity	5
	B. Linear Viscoelastic Theory	7
	C. Relaxation Modulus of Polyethylene Film	12
	D. Nonlinear Viscoelastic Theory	12
	E. Temperature Dependence	16
III	SIMPLIFIED MATERIAL MODELS	23
	A. Background	23
	B. Simplified Analysis	24
IV	PROPOSED NONLINEAR MODEL	41
V	DISCUSSION OF RESULTS	47
	REFERENCES	49

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LIST OF ILLUSTRATIONS

Figure	Title	Page
1	Creep Test	6
2	Stress Relaxation Test	6
3	Typical Presentation of Properties	8
4	Superposition of Stresses	9
5	Generalized Stress-Strain History	9
6	Development of Relaxation Modulus From Constant Rate Tests for 0.5 Mil Polyethylene, T = 23°C	13
7	Time-Temperature Dependence	16
8	Construction of a Stress Relaxation Master Curve of an Amorphous Polymer	19
9	Stress Relaxation Curves of Polyethylene (Hizex) at Various Temperatures	21
10	Comparison of the Predicted and Measured Stresses in Stratofilm Using a Linear Viscoelastic Model	25
11	Uniaxial Stress-Strain Behavior of 1.0 Mil Stratofilm	27
12	Tangent Modulus for 1.0 Mil Stratofilm, Tested at T = -23°C	29
13	Vertical Shift as a Function of Strain, 1.0 Mil Stratofilm, T = -23°C	30
14	Temperature Dependence of h_e	32
15	Derived Linear Relaxation Moduli for Stratofilm	33
16	Derived Master Curve for Relaxation Modulus, Stratofilm	34
17	Master Curve Shift Factors	35
18	Predicted Stress-Strain Curves for Stratofilm	37
19	Measured Response of Stratofilm to Cyclic Strain History	38
20	Predicted Response of Stratofilm to Cyclic Strain History	38

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LIST OF TABLES

Table	Title	Page
1	Proposed Test Matrix for Each Temperature	45

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I. INTRODUCTION

Since the introduction of plastic films in the manufacture of high altitude balloons, scientists have been able to exploit this type of platform in their studies of the atmosphere and space. The engineering community has attempted to keep pace with the demands of the experiments by providing systems that will carry heavier loads to higher altitudes for longer periods of time. Due to the enormous value of these payloads, the reliability of balloon vehicles must be very high. Although many improvements have been made in the areas of film manufacturing, balloon fabrication and operations, there is a need for improved design and analysis techniques. It is believed that the failure of balloons is directly related to the failure of the film in service. Whether these failures are caused by overstress of good quality film or due to flaws or damaged film, the failure is driven by the state of stress in the film. This report is intended to address the nature of the stress state in a film rather than the failure itself.

In order to improve the designs of scientific balloons, the ability to determine the state of stress (and strain) in the balloon throughout the entire flight regime must be developed. This necessitates improved capabilities in three areas: the measurement of actual strains in balloons during real or simulated flights, the ability to calculate the stress field from the measured strains, and the analytical capability to predict the stress and strain in a balloon for all flight conditions. It is the long term aim of the current program at SwRI to satisfy these three requirements.

The most urgent need of these three is the development of a device for accurately measuring stress or strain in balloon film since none currently exists. In a separate paper [1], Rand documents the development of a thin film strain transducer which promises to satisfy this need. The new gage has a low effective modulus so as to not interfere with the strain field in the film, is highly sensitive to strains in the longitudinal direction and relatively insensitive to strains in the transverse direction, and is compensated for the extreme temperature variations common to balloon flights.

With this new transducer, it should be possible to measure strains in the film at various locations along the surface of the balloon. In order to obtain stress values from these strain measurements, an appropriate constitutive equation must be developed which will take into account the interrelationship of stress, strain, temperature and time. This material model would not only be useful in the analysis of flight data but also to improve existing balloon analysis techniques by incorporating a more realistic material model.

The objective of the research documented in this report was to develop a constitutive relationship for polyethylene film which would allow the prediction of stress in the balloon film if the strain history and temperature history were known. It was determined that polyethylene could best be modeled as a nonlinear viscoelastic material, which implied that stress is a function of the material's history. This realization led to a concerted effort

to develop a numerical technique which would allow prediction of the stress in an efficient manner. To this end, an effort was made to combine previously obtained test data with viscoelastic theory to develop a material model of the polyethylene film and a numerical technique which can apply this model to the prediction of stress in the film at any time. The reported work concentrated on the case of uniaxial loading because of its simplicity and availability of test data. Further theoretical and experimental studies are required to develop a working model of the material for uniaxial loading and to generalize the results to the case of biaxial loading.

II. VISCOELASTIC THEORY

During the course of this program, it became apparent that polyethylene film is a highly nonlinear viscoelastic material. The equations needed to describe the material response at a given temperature are complex with numerous functions which must be evaluated through testing. In this section, the general equations used to represent a nonlinear viscoelastic material are developed. Initially, a brief discussion is given of linear viscoelastic theory to acquaint the reader with this area. Data from actual tests on polyethylene film are presented to document the existence and extent of material nonlinearity. Finally, a complex constitutive relationship is developed which will be used to model polyethylene film in the next section.

A. Viscoelasticity

Many solids, including those known as plastics, are referred to as being viscoelastic because they exhibit flow in addition to their elastic characteristics. This flow characteristic is often referred to as "creep" and is readily seen in a simple experiment. In this test, a length of material is "instantaneously" loaded in uniaxial tension with a constant force, as seen in Figure 1a. The resulting strain contains an initial elastic component plus a component which increases with time (creeps) as seen in Figure 1b.

The creep compliance is defined as the time-varying ratio of the strain and stress as given by Equation (1)

$$D(t) = \frac{\varepsilon(t)}{\sigma_c} \tag{1}$$

It should be noted that for an elastic material, strain would be constant for this test, so that the compliance would be a constant.

A dual response of a viscoelastic material is known as stress relaxation, which refers to the response of the material under a constant strain. This response is readily seen in a simple experiment in which a constant strain is applied "instantaneously" to a length of material. The load required to hold the specimen at the constant strain level is seen to decrease with time from the initial load. The strain and resulting stress are seen in Figure 2.

The relaxation modulus is defined as the time-varying ratio of the stress and strain as given in Equation (2)

$$E(t) = \frac{\sigma(t)}{\varepsilon_c} \tag{2}$$

Figure 1. Creep Test

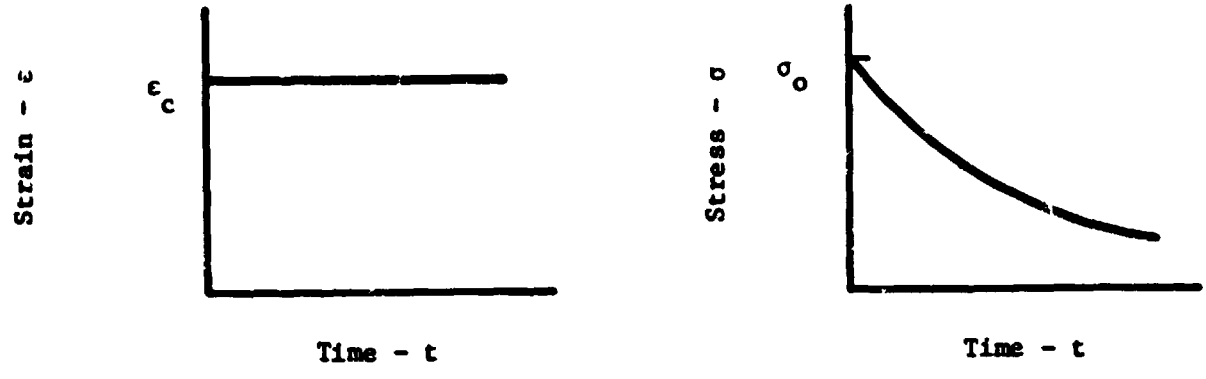


Figure 2. Stress Relaxation Test

For an elastic material, the stress would be constant, and the modulus would be comparable to Young's modulus, a constant. It should also be noted that the compliance, D, and the modulus, E, are reciprocals for an elastic material.

Because of the wide range of times covered in a creep or relaxation test, the creep compliance and relaxation modulus are normally displayed on log-log curves. Examples of these properties are given in Figure 3.

B. Linear Viscoelastic Theory

From the definition of relaxation modulus, Equation (2), if a constant strain, ϵ_c, is applied at time = 0, the stress will be given by the relation

$$\sigma(t) = \epsilon_c \, E(t) \tag{3}$$

For a linear viscoelastic material, if two strains are superimposed and applied at time = 0, the stress will simply be proportionately larger and given by the relation

$$\sigma(t) = [\epsilon_0 + \epsilon_1] \, E(t) \tag{4}$$

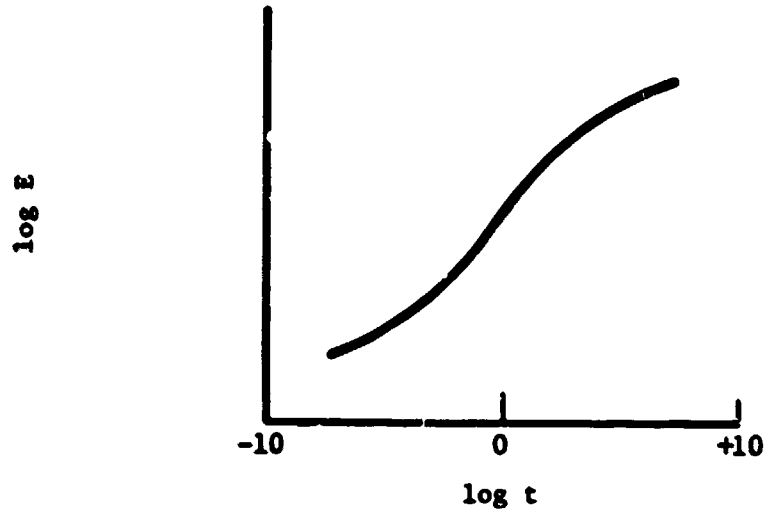
However, for the case in which ϵ_0 is applied at $t = 0$ and ϵ_1 is applied at time, $t = t_1$, (Figure 4), the total stress at some time after t_1 may be approximated by the relationship

$$\sigma(t) = \epsilon_0 \, E(t) + \epsilon_1 \, E(t - t_1) \tag{5}$$

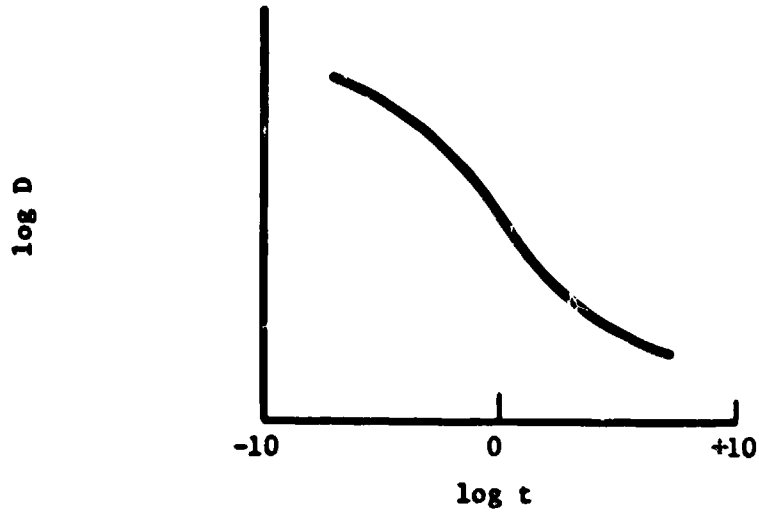
This theory of linear superposition of the two stresses at their elapsed times is referred to as Boltzmann's superposition principle. For a generalized strain history of the form shown in Figure 5, the stress may be approximated by the relationship

$$\sigma(t) = \sum_{i=0}^{N} \epsilon_i \, E(t - \tau_i) \tag{6}$$

For a continuously varying strain history, Equation (6) may be rewritten as



(a) Creep Compliance



(b) Relaxation Modulus

Figure 3. Typical Presentation of Properties

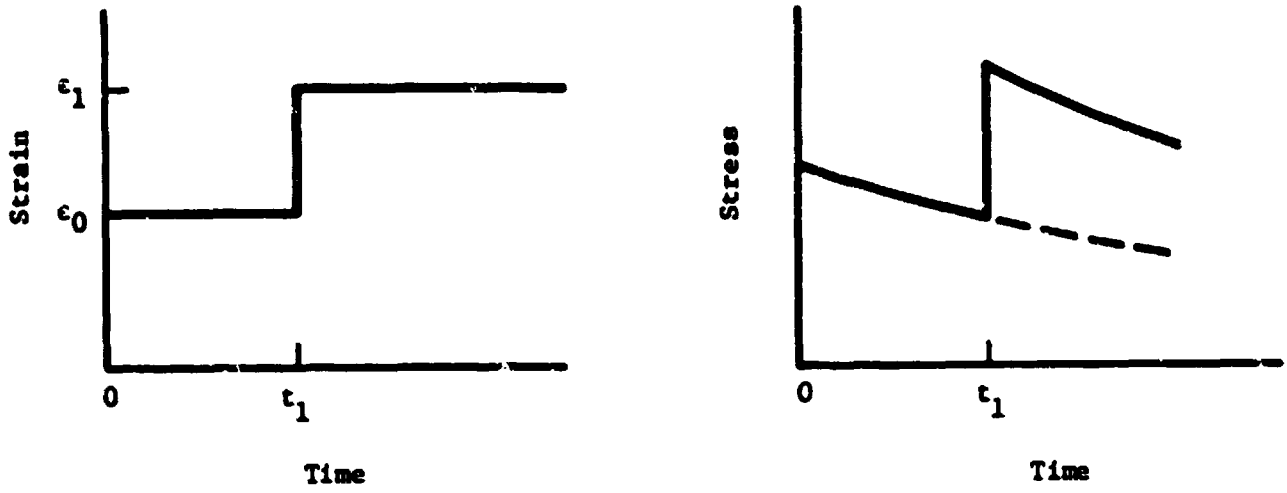


Figure 4. Superposition of Stresses

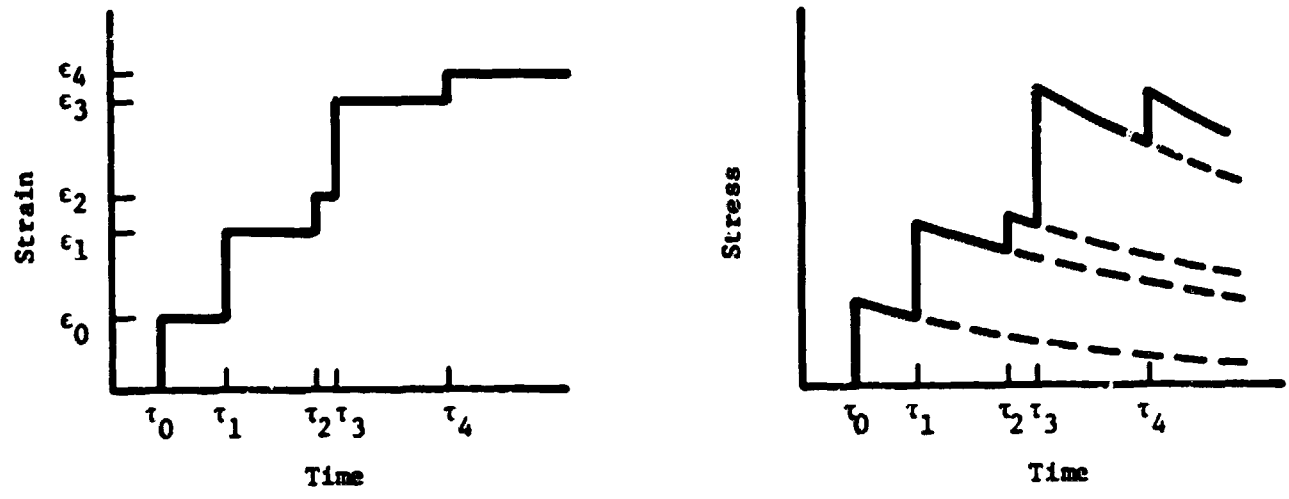


Figure 5. Generalized Stress-Strain History

$$\sigma(t) = \int_0^t E(t - \tau) \frac{\partial \varepsilon}{\partial \tau} d\tau \tag{7}$$

It is now apparent that for a linear viscoelastic material, the stress at any time can be calculated approximately using Equation (7) if the strain history is known and a relationship exists for the relaxation modulus. By a similar derivation, a relationship for strain can be developed corresponding to Equation (7)

$$\varepsilon(t) = \int_0^t D(t - \tau) \frac{\partial \sigma}{\partial \tau} d\tau \tag{8}$$

The relaxation modulus and creep compliance for any material must be determined experimentally. The simplest methods are the creep test (constant stress) and the relaxation test (constant strain) described earlier. The major drawback to these tests is the inability to experimentally apply a finite stress or strain instantaneously. This results in errors in measurement for short times. Thus, these tests are normally used to obtain values of the modulus and compliance applicable for long times only (times greater than a minute). For short time data, a more accurate value of the relaxation modulus of a linear viscoelastic material can be obtained from constant strain-rate tests.

The constant strain-rate test consists of applying a constant rate of elongation and recording the load history. The strain history is taken to be

$$\varepsilon = \begin{cases} o & t < 0 \\ Rt & t \geq 0 \end{cases} \tag{9}$$

where R is the constant rate of strain. The stress history, $\sigma(t)$, is obtained from the loading data. Using this information and Equation (7), one can develop a relationship for the relaxation modulus over a limited time span. Specifically, substituting Equation (9) into Equation (7) gives the result

$$\sigma(t) = R \int_0^t E(t - \tau) \, d\tau \tag{10}$$

By changing the variable of integration

$$v = t - \tau, \; dv = -d\tau$$

Equation (10) takes the form

$$\sigma(t) = R \int_0^t E(v)\, dv \tag{11}$$

This equation can be solved for E(t) by differentiating

$$\frac{1}{R}\, \sigma(t) = \int_0^t E(v)\, dv$$

$$\frac{1}{R}\, \frac{d\sigma}{dt} = E(t)$$

But R dt = dε, resulting in the relationship

$$E(t) = \left. \frac{d\sigma}{d\varepsilon} \right|_{t = \frac{\varepsilon}{R}} \tag{12}$$

Thus, it is seen that the relaxation modulus at the time $t = \varepsilon/R$ is given by the tangent modulus (slope of the stress-strain curve). Using this technique, the relaxation modulus can be obtained over the time-span from fractions of seconds to minutes by conducting multiple constant rate tests at a variety of strain rates.

Once material characterization has resulted in the derivation of the relaxation modulus, then the creep compliance can be derived from it, and vice versa. This can be seen by considering Equation (7) for the case of the strain history, $\varepsilon = D(t)$. For this case, the predicted stress is a unit step stress, such that Equation (7) becomes

$$1 = \int_0^t E(t - \tau) \frac{\partial D}{\partial \tau} d\tau \tag{13}$$

Likewise, Equation (8) may be transformed to the form

$$1 = \int_0^t D(t - \tau) \frac{\partial E}{\partial \tau} d\tau \tag{14}$$

Thus, once E(t) is known, one can theoretically solve either equation in terms of D(t), and vice versa.

C. Relaxation Modulus of Polyethylene Film

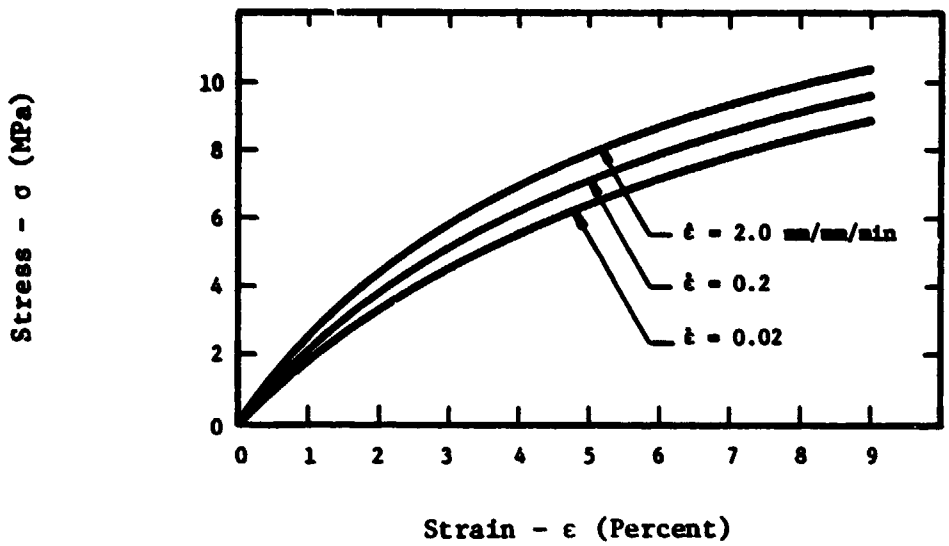
Constant strain rate data was generated for polyethylene film at a variety of rates and temperatures [2]. Curves for the relaxation modulus were generated from this data using Equation (12). Figure 6 displays the stress-strain curves and the derived relaxation modulus for polyethylene film tested at room temperature (T = 23°C) and three different rates of strain. The deviation of the moduli derived for each strain rate in Figure 6b from a single curve was caused by material nonlinearity.

To this point all theoretical considerations have been based on the assumption that the material behaves in a linear viscoelastic manner, i.e., that the relaxation modulus, E, and the creep compliance, D, are functions of time (and temperature) only and not the level of strain. Since both the time dependence and the nonlinearity of the material produce curvature in the constant strain rate data displayed in Figure 6a, the existence and extent of nonlinearity could not be determined by simple inspection of this figure. However, the extent of nonlinearity can be seen plainly in Figure 6b by observing the dashed lines which were obtained by connecting the moduli measured at the same strain levels. The modulus is seen to decrease dramatically with increasing strain.

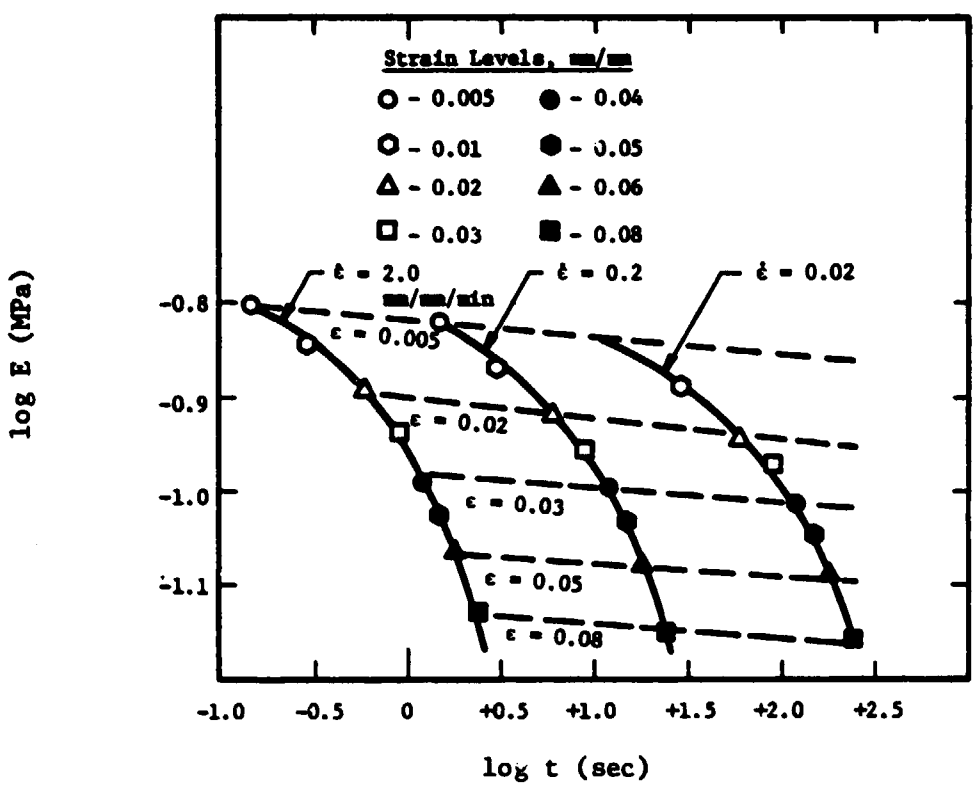
For a nonlinear viscoelastic material, Boltzmann's superposition principle, as represented by Equation (5), is no longer valid. For this reason, our single integral representations of the modulus, Equation (7), and the compliance, Equation (8), are no longer valid.

D. Nonlinear Viscoelastic Theory

Various theories have been developed for representing the constitutive relationships of nonlinear viscoelastic materials [3]. The simplest and most tractable of these appears to be the theory of Schapery [4, 5] which is a modification of the single integral solution represented by



(a) Stress-Strain Curves [2]



(b) Plot of Relaxation Modulus at Various Levels of Strain

Figure 6. Development of Relaxation Modulus from Constant Rate Tests for 0.5 Mil Polyethylene, T = 23°C

Equations (7) and (8) for constant temperature. This theory takes into account the observation that the moduli at high strains in Figure 6b (dashed lines) are the same as those at low values, except for horizontal and vertical translations. Mathematically, Schapery rewrites Equations (7) and (8) for linear viscoelastic response

$$\sigma(t) = E_\infty \, \varepsilon(t) + \int_0^t \Delta E(t - \tau) \frac{\partial \varepsilon}{\partial \tau} \, d\tau \tag{15}$$

$$\varepsilon(t) = D_o \, \sigma(t) + \int_0^t \Delta D(t - \tau) \frac{\partial \sigma}{\partial \tau} \, d\tau \tag{16}$$

where E_∞ represents the equilibrium or final (long time) value of the modulus, D_o represents the initial value of the compliance, and

$$\Delta E(t) = E(t) - E_\infty \tag{17}$$

$$\Delta D(t) = D(t) - D_o \tag{18}$$

Schapery extended these equations to nonlinear response on the basis of his theory of the thermodynamics of irreversible processes. By assuming certain simple forms for free energy and entropy production, he derived the constitutive equations for constant temperature

$$\sigma(t) = h_\infty \, E_\infty \, \varepsilon(t) + h_1 \int_0^t \Delta E(\rho - \rho') \frac{\partial h_2 \varepsilon}{\partial \tau} \, d\tau \tag{19}$$

where

$$\rho = \rho(t) = \int_0^t \frac{dt'}{a_\varepsilon} \qquad (a_\varepsilon > 0) \tag{20}$$

$$\rho' = \rho'(\tau) = \int_0^{\tau} \frac{dt'}{a_\varepsilon} \tag{21}$$

$$a_\varepsilon = a_\varepsilon[\varepsilon(t')]$$

and

$$\varepsilon(t) = g_0 D_o \sigma(t) + g_1 \int_0^t \Delta D(\psi - \psi') \frac{\partial g_2 \sigma}{\partial \tau} d\tau \tag{22}$$

where

$$\psi = \psi(t) = \int_0^t \frac{dt'}{a_\sigma} \qquad (a_\sigma > 0) \tag{23}$$

$$\psi' = \psi'(\tau) = \int_0^{\tau} \frac{dt'}{a_\sigma} \tag{24}$$

$$a_\sigma = a_\sigma[\sigma(t')]$$

In these relationships, h_∞ and h_1 are functions of $\varepsilon(t)$, h_2 and a_ε are functions of $\varepsilon(\tau)$, g_0 and g_1 are functions of $\sigma(t)$, and g_2 and a_σ are functions of $\sigma(\tau)$. For Equation (19), h_∞, h_1, h_2, and a_ε tend toward unity as the strain approaches zero, so that Equation (19) reduces to the linear relationship, Equation (7). For Equation (22), g_0, g_1, g_2, and a_σ tend toward unity as the stress approaches zero, so that Equation (22) reduces to the linear relationship, Equation (8).

Representations of the nonlinear modulus and compliance are obtained by substitution of a constant strain, ε, into Equation (19) and a constant stress, σ, into Equation (22) which yields

$$E_n = \frac{\sigma}{\varepsilon} = h_\infty E_\infty + h_1 h_2 \Delta E \left(\frac{t}{a_\varepsilon}\right) \tag{25}$$

$$D_n = \frac{\varepsilon}{\sigma} = g_0 D_o + g_1 g_2 \Delta D \left(\frac{t}{a_\sigma}\right) \tag{26}$$

where E_n and D_n are the nonlinear modulus and nonlinear compliance respectively.

E. Temperature Dependence

All discussion to this point has assumed that the temperature remains constant. With an increase in temperature the creep and stress-relaxation processes are accelerated, resulting in a large variation of the relaxation modulus and creep compliance with temperature for most polymeric materials. It has been observed for these materials that the shapes of the modulus-time and compliance-time curves (Figure 3) do not change radically, but do suffer a translation parallel to the log t axis.

For a significant number of materials, this shift of property functions parallel to the log t axis without change is sufficiently accurate to be the basis of a thermo-viscoelastic theory. A material which exhibits this behavior is often termed thermorheologically simple and the behavior is termed the time-temperature equivalence. An example of time-temperature equivalence is given in Figure 7.

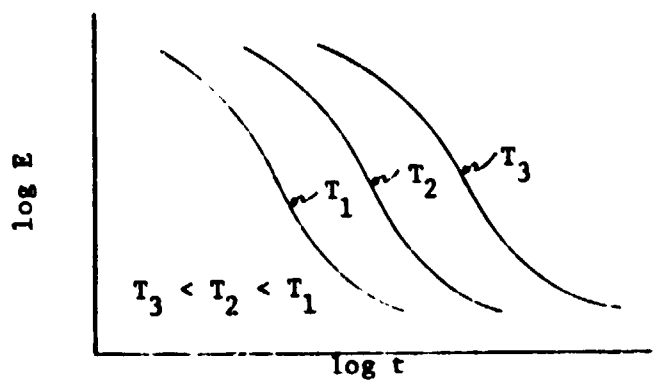


Figure 7. Time-Temperature Dependence

Morland and Lee [6] have shown that for a linearly viscoelastic, thermorheologically simple material, the constitutive relationship of Equation (7) takes the form

$$\sigma(t) = \int_0^t E_m (\xi - \xi') \frac{\partial \epsilon}{\partial \tau} d\tau \tag{27}$$

where

$$\xi = \int_0^t \frac{dt'}{a_T} \text{ - reduced time} \tag{28}$$

$$\xi' = \int_0^\tau \frac{dt'}{a_T} \tag{29}$$

and

$$a_T = a\ \{T(t)\}$$

In Equation (27), $E_m(t)$ is the stress relaxation function at a specific reference temperature, T_m, which is termed the "master" curve; a_T is the shift function such that

$$a_T \Big|_{T = T_m} = 1.0$$

For the case of the temperature held constant at $T \neq T_m$, Equations (28) and (29) can be simplified

$$\xi = \frac{t}{a_T} \tag{30}$$

$$\xi' = \frac{\tau}{a_T} \tag{31}$$

and Equation (27) takes the form

$$\sigma(t) = \int_0^t E_m\left(\frac{t-\tau}{a_T}\right)\frac{\partial\epsilon}{\partial\tau}\,d\tau \tag{32}$$

It should be noted that

$$E\,(t,\,T) = E_m\,(\xi) \tag{33}$$

That is, the relaxation modulus applicable for the temperature T can be obtained from the master curve by shifting the time axis according to Equation (30) or

$$\log \xi = \log t - \log a_T \tag{34}$$

where $\log a_T$ corresponds to the shift along the time axis in Figure 7. By convention

$$a_T = \begin{cases} > 1 & \text{for} \quad T < T_m \\ = 1 & \text{for} \quad T = T_m \\ < 1 & \text{for} \quad T > T_m \end{cases} \tag{35}$$

When the temperature field varies with time, the reduced times, ξ and ξ', are given by Equations (28) and (29) which take into account the entire past history of the temperature field.

A very important feature of the time-temperature equivalence is the ability to use data obtained at different temperatures to derive the relaxation modulus at any given temperature. This is demonstrated in Figure 8 in which relaxation moduli obtained at various temperatures using constant rate tests were used to construct a master curve extending over an extremely large time span.

To this point, the discussion on temperature dependence has assumed that the material is linearly viscoelastic. This is necessary for Equations (27) and (32) to be applicable. However, the time-temperature equivalence has been seen to hold for nonlinear viscoelastic materials as well. Equations (33) and (34) can be applied equally well to the nonlinear equations derived in the last section. In this case, Equations (19) and (22) may be rewritten

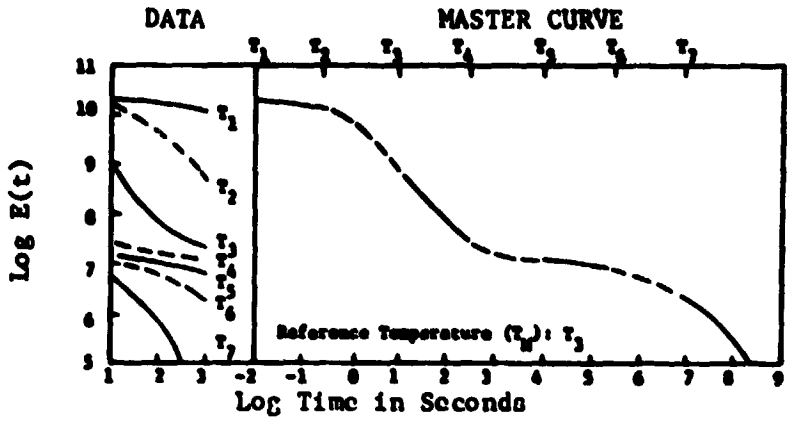


Figure 8. Construction of a Stress Relaxation Master Curve of an Amorphous Polymer (Reference 7)

$$E_N = h_\infty E_\infty + h_1 \int_0^t \Delta E(\beta - \beta') \frac{\partial h_2 \varepsilon}{\partial \tau} d\tau \tag{36}$$

where

$$\beta = \int_0^t \frac{dt'}{a_{\varepsilon T}} \tag{37}$$

$$\beta' = \int_0^\tau \frac{dt'}{a_{\varepsilon T}} \tag{38}$$

$$a_{\varepsilon T} = a_{\varepsilon T} [\varepsilon(t), T]$$

and

$$\varepsilon(t) = g_0 D_o \sigma(t) + g_1 \int_0^t \Delta D(\gamma - \gamma') \frac{\partial g_2 \sigma}{\partial \tau} d\tau \tag{39}$$

where

$$\gamma = \int_0^t \frac{dt'}{a_{\sigma T}} \tag{40}$$

$$\gamma' = \int_0^\tau \frac{dt'}{a_{\sigma T}} \tag{41}$$

$$a_{\sigma T} = a_{\sigma T}\,[\sigma(t),\ T]$$

The major change in these equations was the addition of temperature dependence to a_ε and a_σ defined earlier.

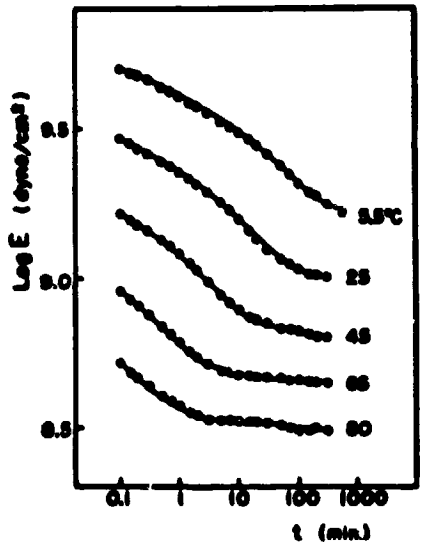
Finally, it has been noted [8] that the condition of time-temperature equivalence is not a universally valid rule. In order to increase its range of applicability, attempts have been made to model data by including a magnification factor (a vertical shift of the curves parallel to the logarithmic modulus axis) in addition to the horizontal shift already discussed.

Such horizontal and vertical shifts have been used previously [8] to develop master curves for several types of polyethylene film. The master curve for Hizex, displayed in Figure 9b, was formed by shifting the stress relaxation curves of Figure 9a. The shift factors were

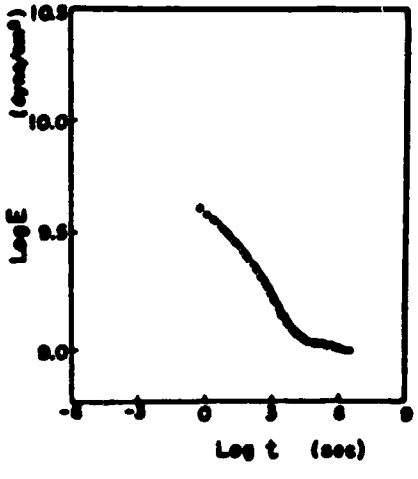
horizontal $\log a_T = -0.035\ (T - 20°C)$

vertical $\log h_T = -0.0083\ (T - 20°C)$

The presence of the vertical shift implies that this polyethylene film (Hizex) is not a thermorheologically-simple material. However, the techniques presented above appeared to be sufficient for developing a realistic master curve for this material.



(a)



(b) Master Curve

Figure 9. Stress Relaxation Curves of Polyethylene (Hizex) at Various Temperatures [8]

III. SIMPLIFIED MATERIAL MODELS

The purpose of this ongoing program is to develop the relationships for characterizing the response of balloon film under load. Of initial importance is a relationship which would allow the calculation of stress for a known strain history. This is needed to process data attained using strain gages during balloon material tests and actual flights.

Because of the complexities involved in developing a nonlinear model of film which accurately predicts response over a wide range of temperature and loading conditions, emphasis has been placed on developing simplified models initially. In this section, a review of previous work will be given. An explanation of a simplified analysis developed in this program will also be given. This simplified model was developed to lend insights into the nonlinear processes and to demonstrate the feasibility of properly modeling film response with a nonlinear model. Emphasis in present work has been on model development for uniaxial loading while biaxial effects will be considered in later work.

A. Background

The mechanical response of polyethylene has been studied under a variety of loading and temperature conditions by a number of investigators. Most notable has been the data collected by Alexander and Weissman [9] at Stevens Institute of Technology, Okamoto [10], and Webb [2] at Texas A&M University. Unfortunately, raw data cannot be effectively utilized in any logical analysis procedure. Alexander characterized the film as a linearly-elastic, orthotropic material with material properties evaluated as a function of only temperature. However, Webb [2] experimentally showed that the mechanical properties are a function of time as well as temperature. This suggested that the material is viscous in nature, although not necessarily viscoelastic.

It was suggested by Schapery [11] that if the data are restricted to very small strains, then linear viscoelastic characterization might be appropriate. Thus, Webb [2] conducted an extensive effort to characterize polyethylene film as a linear viscoelastic material. Several simplifying techniques were incorporated into his analyses including a linear time-temperature shift, a simple power law representation of the tensile modulus and the assumption that the modulus and compliance were reciprocals. In this program, initial work concentrated on developing a relationship for the creep compliance which was a function of both time and temperature. The compliance was chosen since it can be developed from creep test data, which is relatively easy to generate and does not require extensive test equipment. The relaxation modulus was then assumed to be the reciprocal of the derived creep compliance. The resulting relationship for the modulus was

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$$E(t) = 20{,}000 \left(\frac{t}{a_T}\right)^{-0.1} \tag{42}$$

where

$$\log a_T = -0.116\ T + 2.668 \tag{43}$$

and

E(t) is in psi
t is in minutes
T is in °C.

This relationship was based on tests of three thicknesses of balloon film (0.5, 0.7, and 1.0 mil) with a trade name of Stratofilm.

As a check of this relationship, constant strain rate tests were conducted and the results were checked against predictions made using Equation (42) and the convolution integral, Equation (7). For this case, the convolution integral took the form

$$\sigma(t) = R \int_0^t 20{,}000 \left(\frac{t - \tau}{a_T}\right)^{-0.1} d\tau$$

Constant rate data was generated for three temperatures and three rates. Results of the predicted and measured stresses are given in Figure 10.

A poor correlation was found between the measured and predicted stresses, especially at higher strains. The reasons for this are rather straightforward: the simple power law representation of the material was inadequate, material nonlinearities of the material were not considered, temperature shifting was overly simplified, and the assumption that the compliance and modulus are reciprocals was invalid.

B. Simplified Analysis

In order to properly model the response of polyethylene film, an expression for the relaxation modulus was developed which took the form of Equation (25)

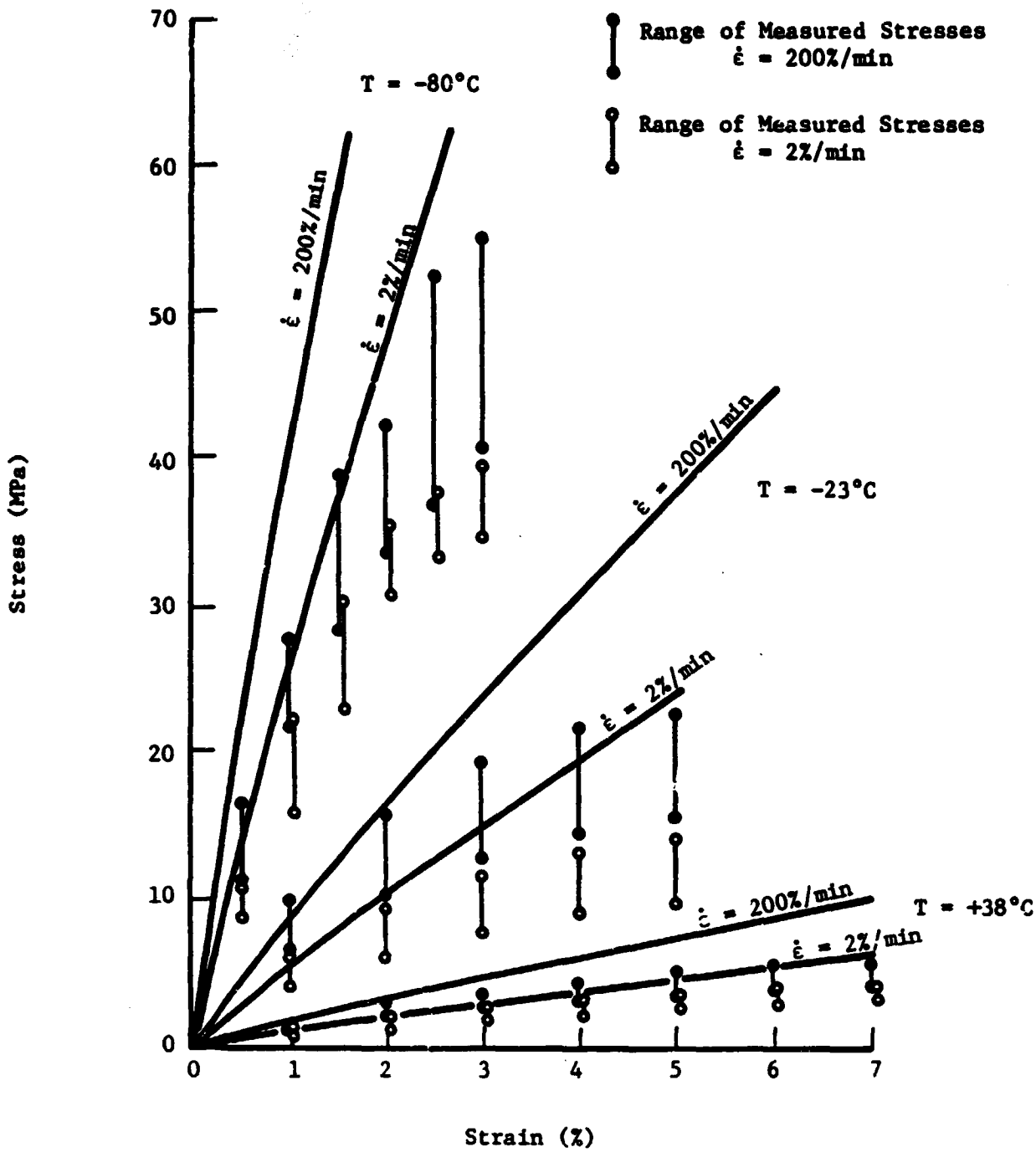


Figure 10. Comparison of the Predicted and Measured Stresses in Stratofilm Using a Linear Viscoelastic Model [2]

$$E_n = h_\infty E_\infty + h_1 h_2 \Delta E \left(\frac{t}{a_\varepsilon}\right)$$

with four functions, h_∞, h_1, h_2, and a_ε, which must be evaluated experimentally for any given temperature. An extensive test program is required to accurately evaluate these functions over the range of temperatures of interest. Since data from this type of experimental program does not currently exist, an effort was made to use existing data to develop a simplified expression for the nonlinear modulus. Such an expression, though incapable of modeling the response of the material under all conditions, would be useful in evaluating the effects of material nonlinearities on the prediction of stress and strain.

One approach to developing a simplified expression for the modulus is to modify Equation (7) such that

$$\sigma(t) = \int_0^t E_n (t - \tau) \frac{\partial \varepsilon}{\partial \tau} d\tau \tag{44}$$

where the linear modulus has been replaced by its nonlinear counterpart, which has the form

$$E_n(t) = h_e E \left(\frac{t}{a_e}\right) \tag{45}$$

This simplified relationship is similar to Equation (25) for the case of $h_\infty = h_1 h_2 = h_e$ and $a_\varepsilon = a_e$. Equation (45) can be rewritten

$$\log E_n(t) = \log h_e + \log E \left(\frac{t}{a_e}\right) \tag{46}$$

If experimentally obtained modulus data is plotted against t on double-logarithmic paper, curves at different strain levels can be superposed by translation along the two axes. The amount of vertical shift corresponds to log h_e and the amount of horizontal shift to log a_e. This process is similar to that explained in Section II for temperature shifting.

The only data currently available for evaluating the two nonlinear coefficients in Equation (45) is the constant strain rate data obtained by Webb [2]. The stress-strain data given in Figure 11 is for 1.0 mil Stratofilm which was tested in the transverse direction. Information concerning

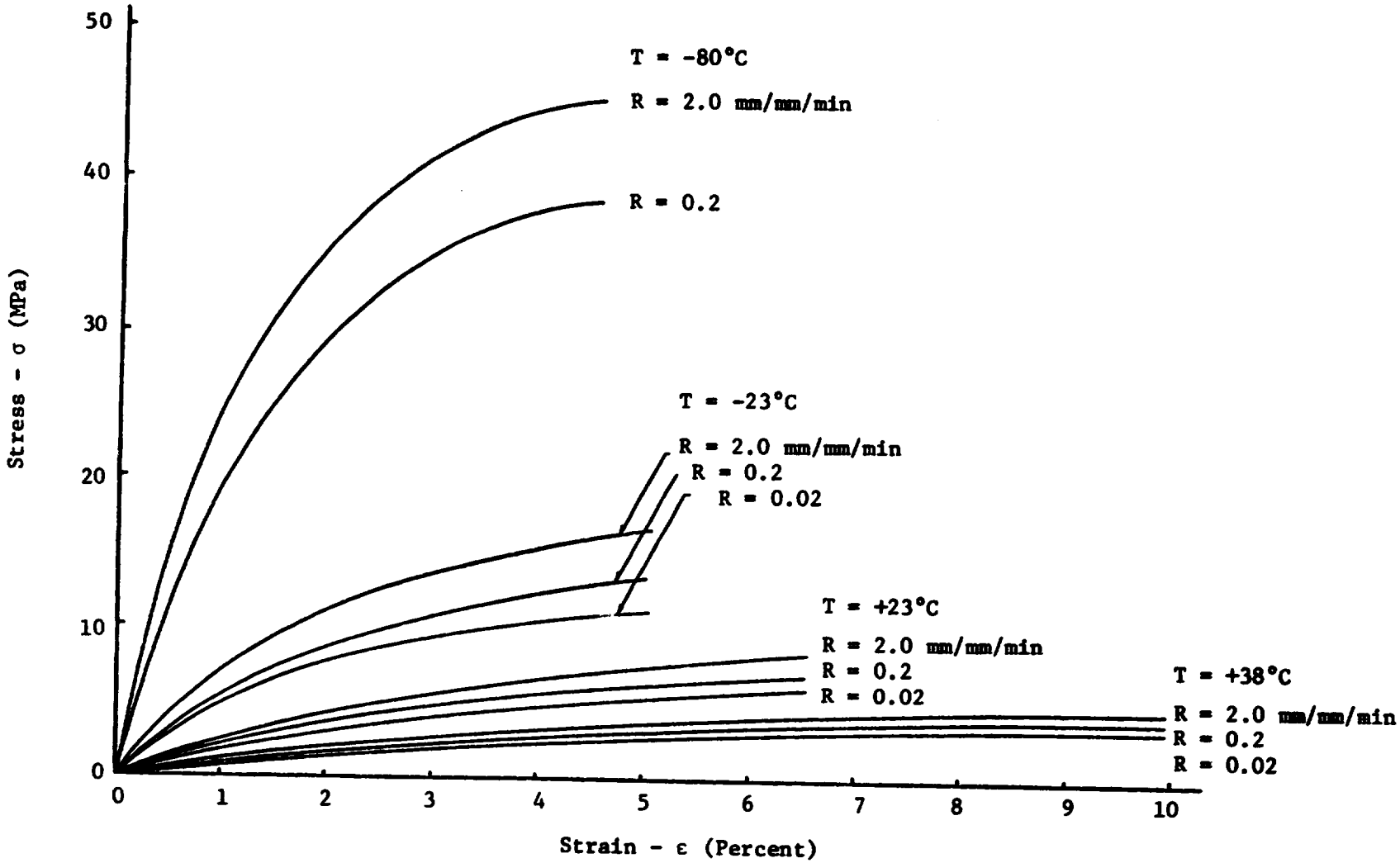


Figure 11. Uniaxial Stress-Strain Behavior of 1.0 Mil Stratofilm

the relaxation modulus can be obtained by measuring the slope of the stress-strain curve. According to Equation (12), the relaxation modulus at the time $t = \varepsilon/R$ is given by the measured tangent modulus (slope of the stress-strain curve). The measured values of the tangent modulus obtained from the data in Figure 11 for T = -23°C is given in Figure 12. A separate curve was generated for each strain rate.

It should be noted that these curves were generated from data for which the strain is constantly changing, whereas the relaxation modulus is defined as the ratio of stress to strain for a constant strain. Thus, to obtain curves for the relaxation modulus from tangent modulus curves, points on each curve in Figure 12 for a given strain must be connected. As seen in the figure, dashed lines have been added which connect the points of constant strain. These dashed lines represent the curves for relaxation modulus at strains of 1, 2, 3, and 4 percent. The fact that these curves are not superposed indicates that the modulus is a function of strain, and therefore, nonlinear. As the strain level decreases, each of the tangent modulus curves tend toward a single curve, which represents the apparent linear relaxation modulus.

Looking again at Equation (46), the linear modulus is represented by E(t) and the dashed lines can be superposed on the linear modulus curve by vertical and horizontal translations, with the vertical translation represented by log h_e and the horizontal translation by log a_e. In order to simplify the model being developed, a decision was made that only vertical translations would be used to shift the data, that is, $a_e = 1$. A plot of the measured vertical translations are given in Figure 13. The expression for h_e derived from this curve took the form

$$\frac{d \log h_e}{d\varepsilon} = -17.2$$

or

$$h_e = e^{-17.2\varepsilon} \tag{47}$$

This relationship, along with Equation (45), is sufficient to model the material if the temperature remains at -23°C. However, a relationship is needed which can be used to describe the response of film to the entire range of temperatures experienced in balloon flight. Such a relationship can be developed by deriving relationships similar to Equation (47) for a range of temperatures and then developing a master curve as described in Section II. The horizontal and vertical shifting done in constructing the master curve will result in Equation (45) taking the form

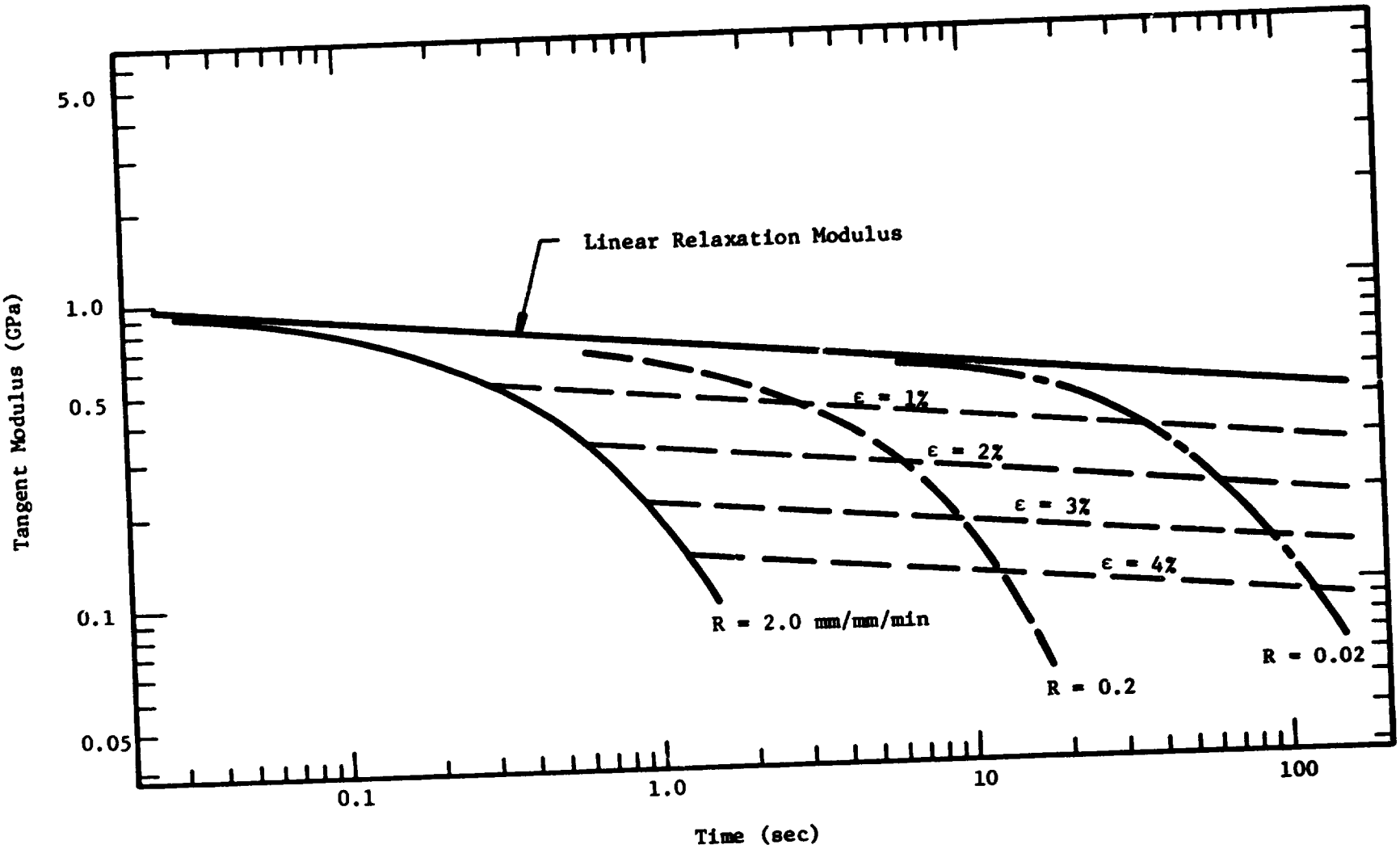


Figure 12. Tangent Modulus for 1.0 Mil Stratofilm, Tested at T = -23°C

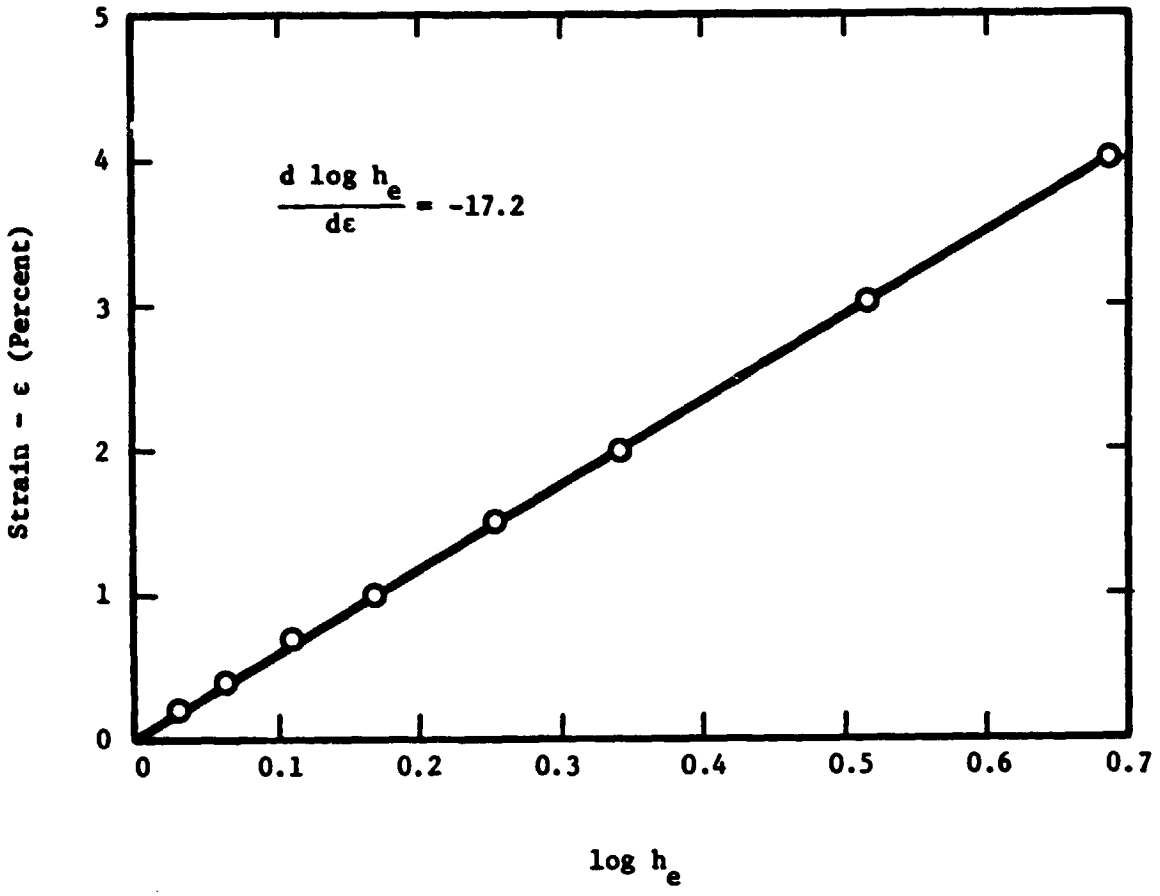


Figure 13. Vertical Shift as a Function of Strain, 1.0 Mil Stratofilm, T = -23°C

$$E_n(t) = h_T\, h_e\, E\left(\frac{t}{a_T}\right) \tag{48}$$

where it has been assumed that $a_e = 1$ for all temperatures.

Expressions similar to Equation (47) were derived for much of the data generated by Webb [2], and is displayed in Figure 14. From this figure, an expression for h_e as a function of temperature was found

$$h_e = e^{(-27.63 + 0.394\ T)\ \varepsilon} \tag{49}$$

with T in °C.

Curves similar to the linear modulus of Figure 12 were developed for several series of tests, including 0.5 mil film tested in the machine direction and 1.0 mil film tested in the machine and transverse directions. These apparent linear moduli are plotted in Figure 15 for the four test temperatures. In a process similar to that demonstrated in Figure 8, a master curve was constructed from these curves, and is presented in Figure 16. Thus, the linear relaxation moduli obtained at various temperatures over a short time span were used to construct a master curve extending over an extremely large time span. A modified power law was developed which adequately represents the master curve and which has the form

$$E(t) = 0.0069\ \text{GPa} + \frac{2.30\ \text{GPa}}{\left(1 + \dfrac{t}{2.816 \times 10^{-7}}\right)^{0.094}} \tag{50}$$

with t in seconds.

The amount of horizontal and vertical shifting which was required to obtain Figure 16 is plotted in Figure 17. From this figure, the expressions for a_T and h_T were found to be approximately

$$a_T = e^{(1.635 - 0.0716\ T)} \tag{51}$$

$$h_T = e^{(0.409 - 0.0179\ T)} \tag{52}$$

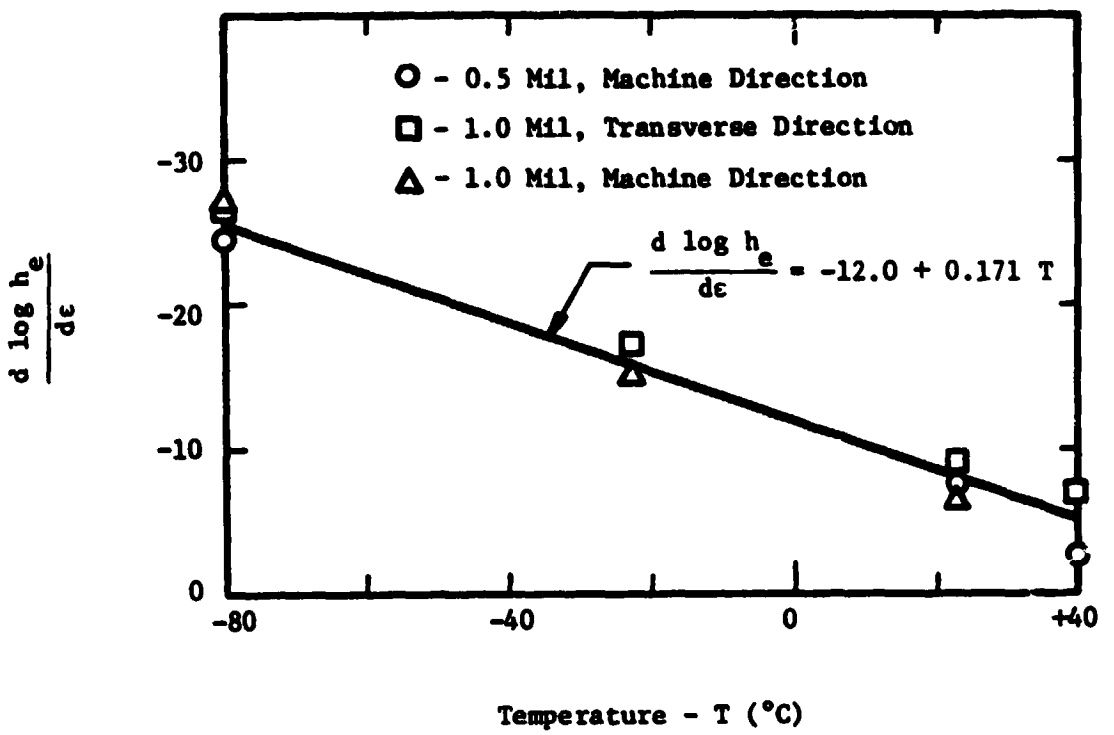


Figure 14. Temperature Dependence of h_e

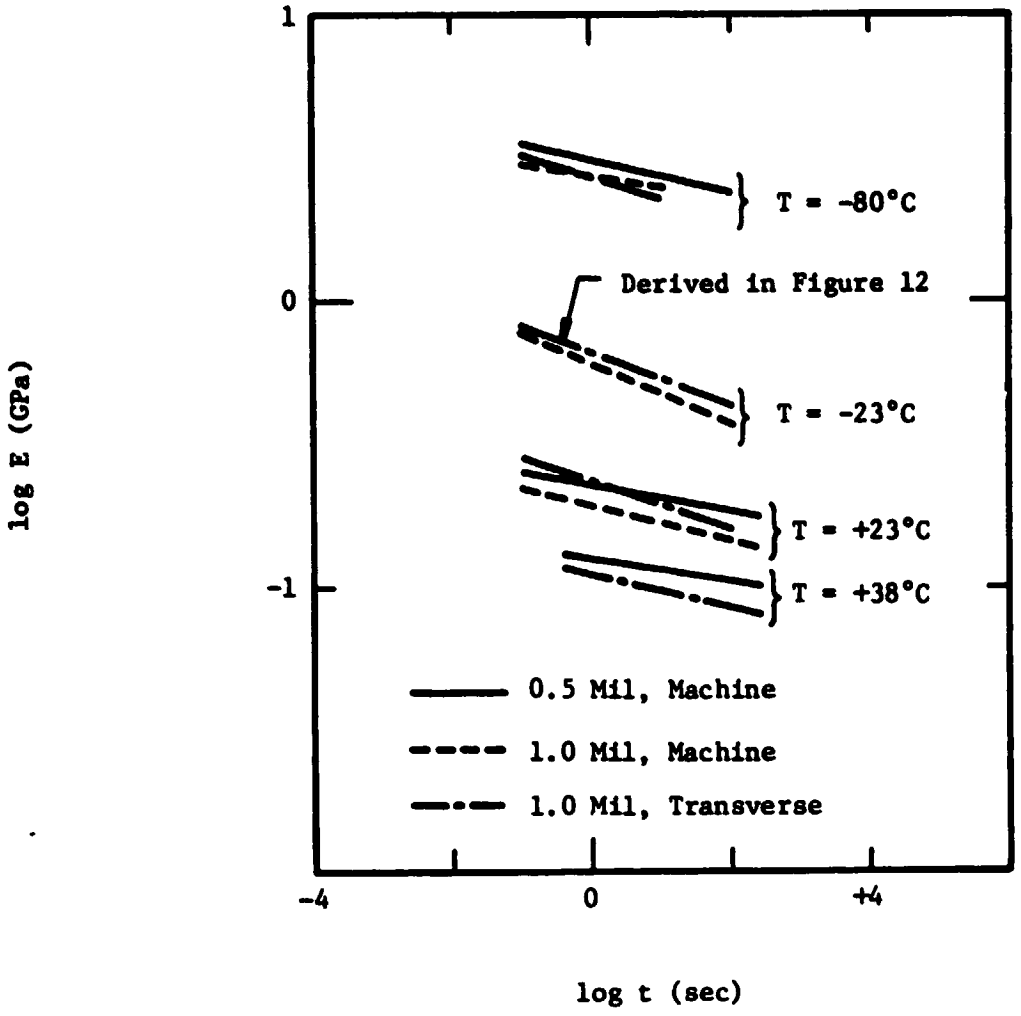


Figure 15. Derived Linear Relaxation Moduli for Stratofilm

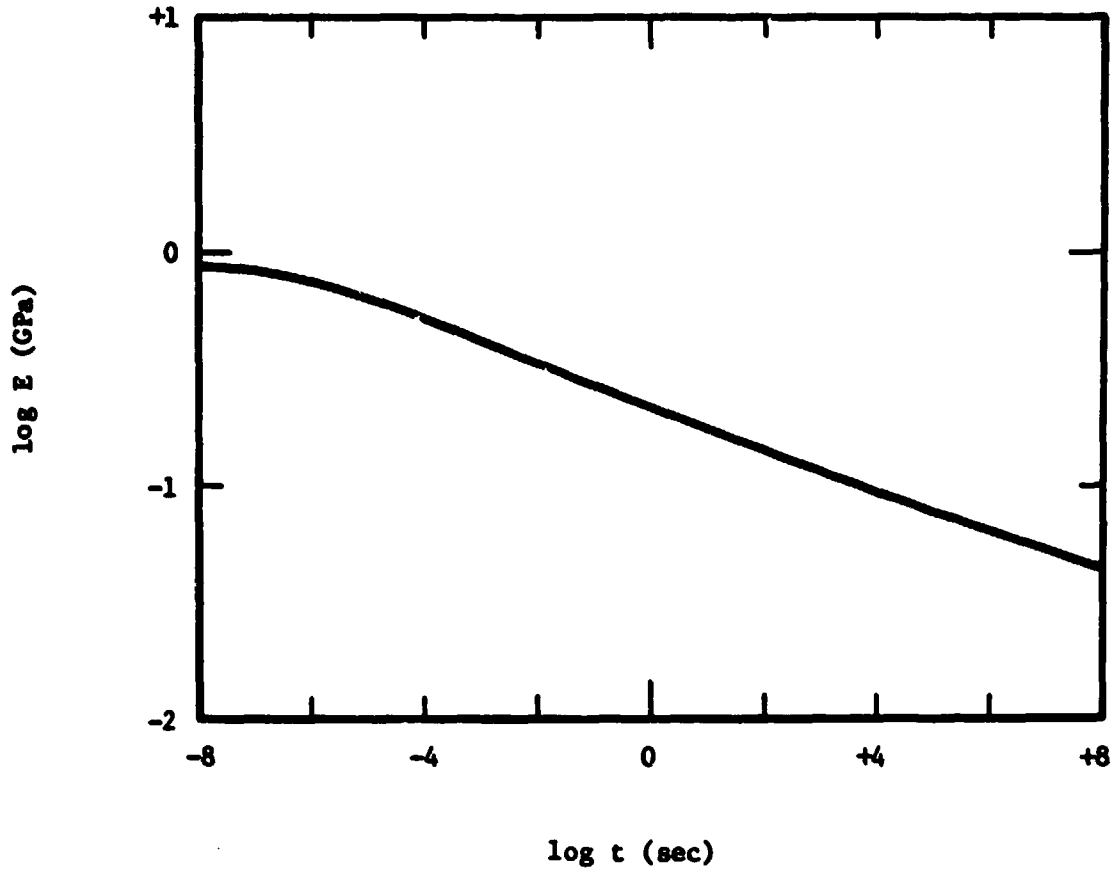
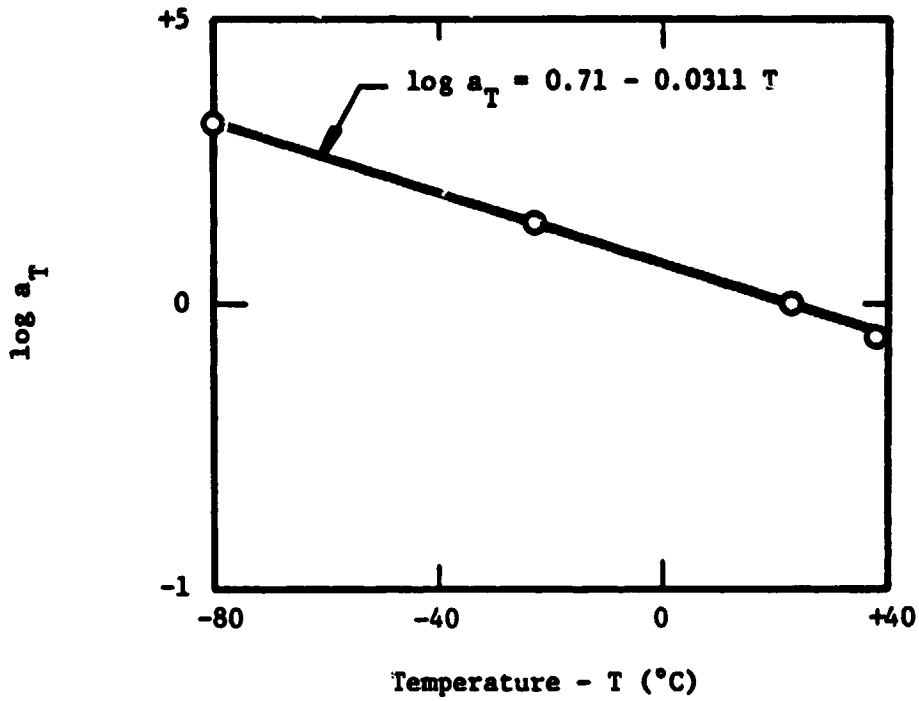
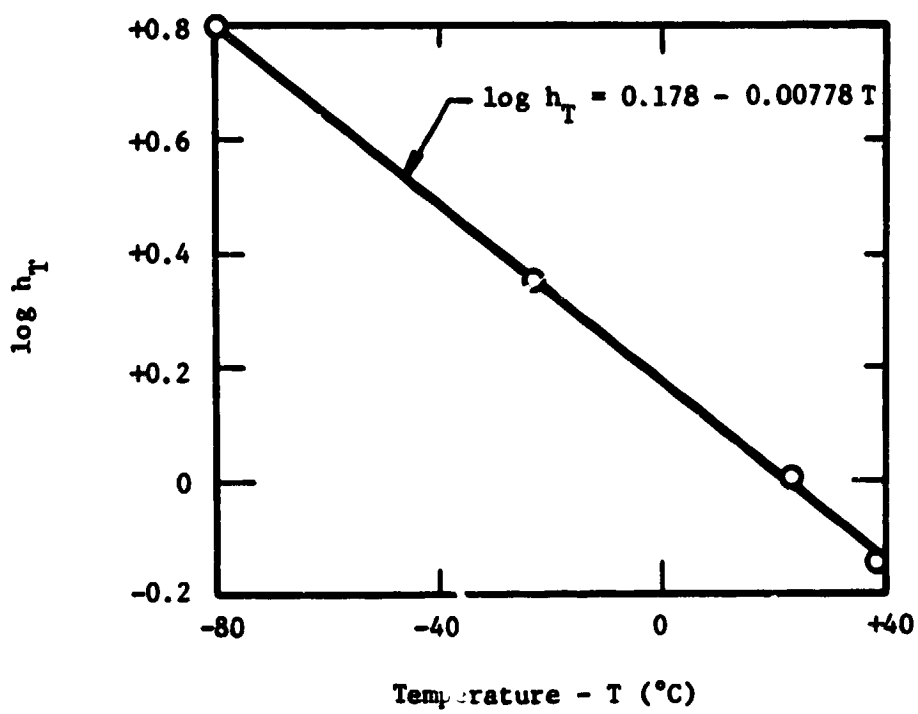


Figure 16. Derived Master Curve for Relaxation Modulus, Stratofilm



(a) Horizontal Shifts



(b) Vertical Shifts

Figure 17. Master Curve Shift Factors

Now, the expressions given in Equations (48) through (52) are sufficient to describe the response of the film over the entire range of temperatures and times which are experienced in balloon flights.

As a partial check of the validity of the simple model just developed, a decision was made to predict the stress-strain curves which had been used initially as input data in the derivation of the model. This was done by substituting the simplified nonlinear model represented by Equations (48) through (52) into the relationship for stress as a function of strain, Equation (44). Equation (44) was numerically integrated for twelve cases comprised of tests at four temperatures at each of three strain rates. The results of these calculations are given in Figure 18. Comparisons of these results with some of the input data given in Figure 11 shows that the nonlinear model does a good job qualitatively of duplicating the stress-strain curves over the range of rates and temperatures involved. These predictions greatly contrast those made using the linear model developed by Webb [2] and displayed in Figure 10. The simplified nonlinear model closely models the decrease in modulus with increasing strain which is almost completely absent with the linear model.

Although the simple nonlinear model shows good qualitative agreement with the limited data, it is still inadequate as a model for quantitative calculations, as typified by its inability to duplicate the measured response at T = +38°C. Further work with this model has demonstrated its inability to properly model the case of cyclic loading, such as is present in an actual balloon flight.

In a simple cyclic loading test, a thin tensile film specimen was subjected to the following strain history:

$$\dot{\varepsilon} = \begin{cases} +2 \text{ mm/mm/min} & 0 \leq t < 5 \text{ min} \\ -2 \text{ mm/mm/min} & 5 \text{ min} \leq t < 7 \text{ min} \\ +2 \text{ mm/mm/min} & 7 \text{ min} \leq t < 12 \text{ min} \\ -2 \text{ mm/mm/min} & 12 \text{ min} \leq t < 14 \text{ min} \\ +2 \text{ mm/mm/min} & 14 \text{ min} \leq t < 18 \text{ min} \end{cases}$$

The temperature was held constant at T = +23°C. Typical results of this test are given in Figure 19. The predicted response of the film using the simple nonlinear model is given in Figure 20. An examination of the predictions show that the simple nonlinear model does not properly model the release process. This is probably caused by the inability to model all aspects of the material's history dependence with a single nonlinear function, h_e.

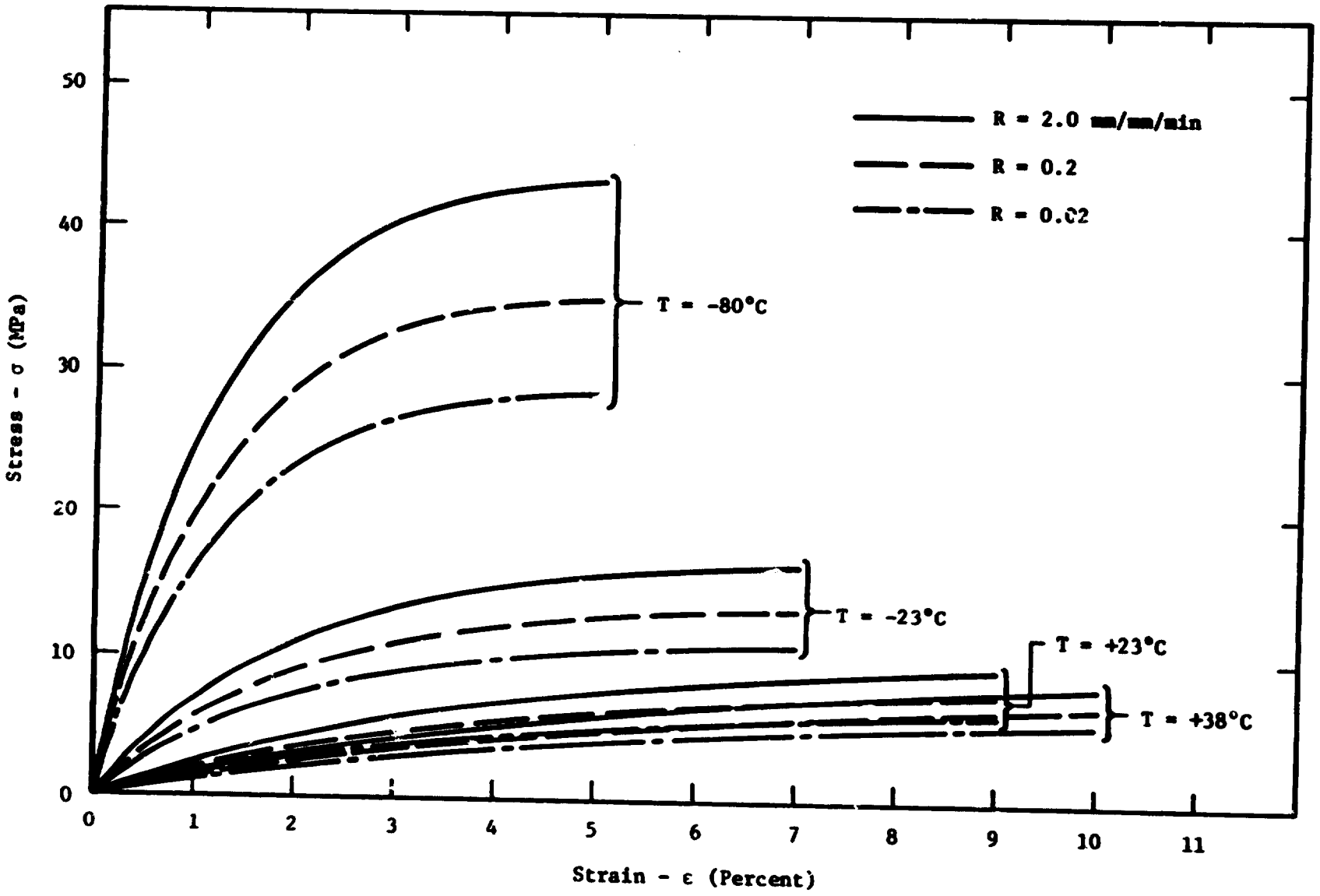


Figure 18. Predicted Stress-Strain Curves for Stratofilm

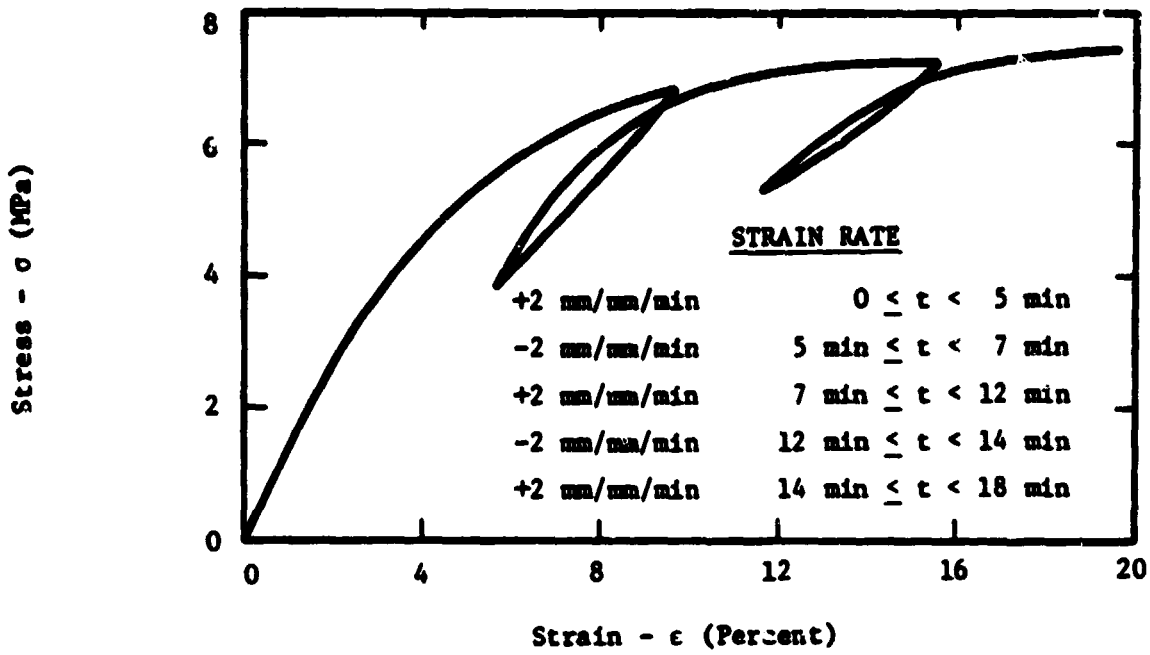


Figure 19. Measured Response of Stratofilm to Cyclic Strain History

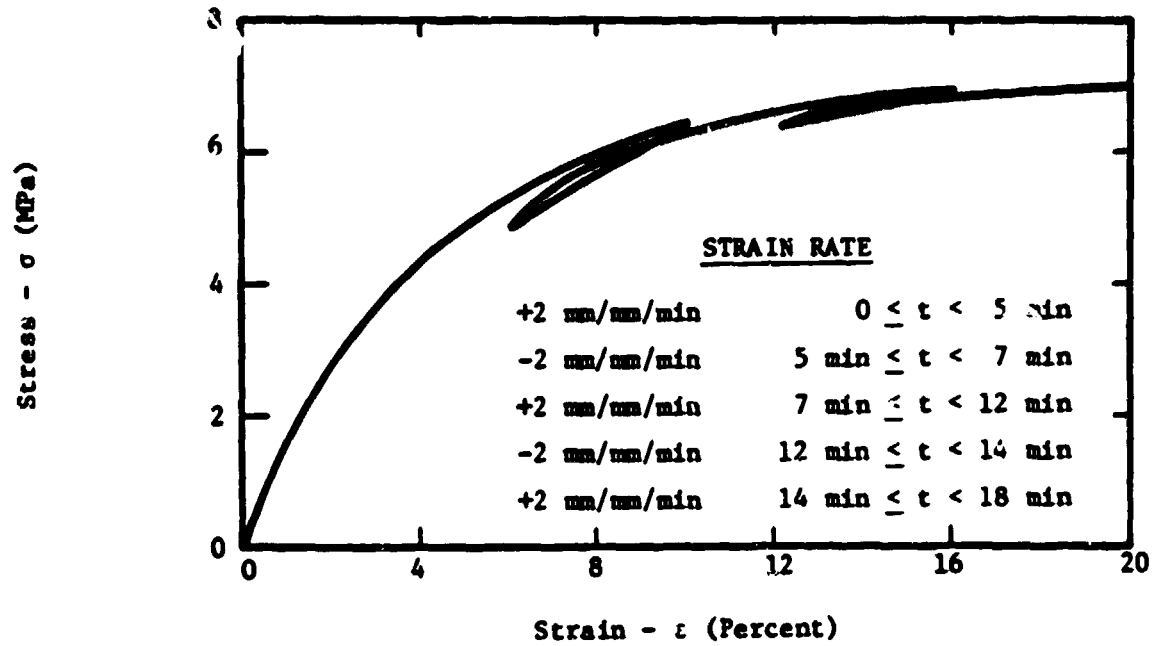


Figure 20. Predicted Response of Stratofilm to Cyclic Strain History

In the next section, four nonlinear functions are included in the model for the relaxation modulus. These additional terms should allow the model to more properly duplicate the actual response of balloon film to the flight environment.

IV. PROPOSED NONLINEAR MODEL

In order to properly model polyethylene film, a more complex nonlinear model must be used than the simple model considered in Section III. The desired model chosen for this study was based on the Schapery thermodynamic theory [4, 5]. The constitutive equation for the stress in terms of the strain history was given in Equation (19)

$$\sigma(t) = h_\infty E_\infty \varepsilon(t) + h_1 \int_0^t \Delta E \, (\rho - \rho') \frac{\partial h_2 \varepsilon}{\partial \tau} d\tau$$

with a corresponding nonlinear relaxation modulus given by Equation (25)

$$E_n(t) = \frac{\sigma(t)}{\varepsilon} = h_\infty E_\infty + h_1 h_2 \Delta E \left(\frac{t}{a_\varepsilon}\right)$$

The functions h_∞, h_1, h_2, and a_ε are dependent upon the strain. More specifically, h_∞ and h_1 are seen to be functions of the current strain, $\varepsilon(t)$, and h_2 and a_ε are functions of the entire strain history, $\varepsilon(\tau)$. It should be noted that for a linear material, $h_\infty = h_1 = h_2 = a_\varepsilon = 1$, and Equation (25) reduces to the linear relationship, Equation (17)

$$E(t) = E_\infty + \Delta E(t)$$

In order to evaluate Equation (25) for a given material, an extensive test program is required. In this process, relaxation tests are conducted at a variety of temperatures. For each temperature, a value is obtained for E_∞, an expression is developed for the transient linear modulus, $E(t)$, as a function of time, and expressions are generated for h_∞, h_1, h_2, and a_ε as functions of strain.

In order to generalize Equation (25) for all temperatures of interest, expressions are developed for the four nonlinear terms as functions of temperature as well as strain. This process is similar to that used to evaluate the nonlinear function, h_e, of the simplified model as seen in Figure 13, with resulting relationships similar in form to Equation (49). Generalization of E_∞ and $\Delta E(t)$ in terms of temperature is accomplished in the manner discussed in Section II. The linear modulus, $E_\infty + \Delta E(t)$, is plotted for each different temperature, and then shifted vertically and horizontally to form a master curve. The vertical shift is used to develop an expression, η, and

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the horizontal shift is used to develop a_T. The resulting expression for the linear portion of the modulus takes the form

$$E(t, T) = h_T E_\infty + h_T \Delta E \left(\frac{t}{a_T}\right) \tag{53}$$

The full expression for the nonlinear modulus then has the form

$$E_n(t, T) = h_T h_\infty E_\infty + h_T h_1 h_2 \Delta E \left(\frac{t}{a_\varepsilon a_T}\right) \tag{54}$$

By combining several terms, Equation (54) may take the form of Equation (25)

$$E_n(t, T) = h_\infty' E_\infty + h_1' h_2 \Delta E \left(\frac{t}{a_\varepsilon'}\right) \tag{55}$$

where $h_\infty' = h_T h_\infty$, $h_1' = h_T h_1$, and $a_\varepsilon' = a_T a_\varepsilon$.

The key to developing this relationship for the nonlinear modulus is the evaluation of the four functions h_∞, h_1, h_2, and a_ε at each of the temperatures since the remainder of the process is similar to that used in developing the simple model of Section III. These functions can be evaluated empirically through the use of a carefully conducted series of relaxation tests at a variety of temperatures. This process will not be explained.

Initially, a series of one-step relaxation tests should be conducted at a variety of strain levels and temperatures. The strain histories will be of the form

$$\varepsilon = \begin{cases} 0 & t < 0 \\ \varepsilon^a & 0 \leq t < t_a \end{cases}$$

For these tests, the proposed nonlinear relaxation modulus has the form

$$E_n(t) = \frac{\sigma(t)}{\varepsilon^a} = h_\infty E_\infty + h_1 h_2 \Delta E \left(\frac{t}{a_\varepsilon}\right) \tag{56}$$

for all tests at a given temperature. Now, the transient component of the modulus is seen to be

$$\Delta E_n(t) = E_n(t) - h_\infty E_\infty$$
$$= h_1 h_2 \Delta E \left(\frac{t}{a_\varepsilon}\right) \tag{57}$$

This relation may also be written

$$\log \Delta E_n(t) = \log h_1 h_2 + \log \Delta E \left(\frac{t}{a_\varepsilon}\right) \tag{58}$$

If the results of several relaxation tests conducted at different levels of strain but the same temperature are plotted on a double logarithmic graph, they should appear similar to the dashed lines of Figure 12. Ideally, one of the tests should have been conducted at sufficiently low strain to assure linear response. The results of the linear test would then correspond to the linear curve in Figure 12. If the relaxation tests were conducted for a sufficiently long time, values for the equilibrium moduli should be found from these curves. For the linear curve, this value corresponds to E_∞, and for the other curves, it will be $h_\infty E_\infty$. Thus, relationships can be obtained for both E_∞ and h_∞ for each temperature tested. A check of E_∞ can be made by conducting a long term creep test with sufficiently low stress to insure linear response. The measured equilibrium creep value, D_∞, should be the reciprocal of E_∞.

By subtracting the equilibrium values of the moduli, curves for the transient portions of the moduli, $\Delta E_n(t)$, can be obtained. When these are plotted against time, t, on double logarithmic paper, it should be possible to superpose all of the curves by translating them along the two axes. This process was followed in the last section to obtain values for h_e and a_e. In the present case, the horizontal shift will be equal to log a_e and the vertical shift to log $h_1 h_2$. Thus, if this process is successfully followed, values can be obtained for a_ε and the product $h_1 h_2$ for a given temperature.

In order to obtain separate values for h_1 and h_2, a series of two-step relaxation tests must be conducted at various temperatures. For these tests, the strain histories will be

$$\varepsilon = \begin{cases} 0 & t < 0 \\ \varepsilon^a & 0 \leq t < t_a \\ \varepsilon^b & t_a \leq t < t_b \end{cases}$$

Substituting this strain history into Equation (19) results in the following relationships [12]

$$0 \leq t < t_a$$

$$\sigma = \varepsilon^a \left[h_\infty{}^a E_\infty + h_1{}^a h_2{}^a \Delta E \left(\frac{t}{a_\varepsilon{}^a} \right) \right] \tag{59}$$

and

$$t_a \leq t < t_b$$

$$\sigma = h_\infty{}^b E_\infty \varepsilon^b + h_1{}^b \left[h_2{}^a \varepsilon^a \Delta E \left(\frac{t_a}{a_\varepsilon{}^a} + \frac{t - t_a}{a_\varepsilon{}^b} \right) \right. \tag{60}$$

$$\left. + \left(h_2{}^b \varepsilon^b - h_2{}^a \varepsilon^a \right) \Delta E \left(\frac{t - t_a}{a_\varepsilon{}^b} \right) \right]$$

which may be rearranged to obtain

$$\sigma = \left[h_\infty{}^b E_\infty + h_1{}^b h_2{}^b \Delta E \left(\frac{t - t_a}{a_\varepsilon{}^b} \right) \right] \varepsilon^b$$

$$- \left(\frac{h_1{}^b}{h_1{}^a} \right) h_1{}^a h_2{}^a \left[\Delta E \left(\frac{t - t_a}{a_\varepsilon{}^b} \right) - \Delta E \left(\frac{t_a}{a_\varepsilon{}^a} + \frac{t - t_a}{a_\varepsilon{}^b} \right) \right] \varepsilon^a \tag{61}$$

The integrations used to derive Equations (59) and (60) were carried out recognizing that $\partial h_2\varepsilon/\partial\tau$ is zero except at $t = 0$ and $t = t_a$.

If one-step relaxation tests were conducted for the strains ε^a and ε^b, then values should already be known for E_∞, $h_\infty{}^a$, $h_\infty{}^b$, $a_\varepsilon{}^a$, $a_\varepsilon{}^b$, and the products $h_1{}^a\ h_2{}^a$ and $h_1{}^b\ h_2{}^b$. Thus, all terms are known in Equation (61) except for the ratio $(h_1{}^b/h_1{}^a)$ which can then be obtained by matching Equation (61) to the two-step data at a convenient point ($t > t_a$). At least one test must be run in which ε^b was chosen to be sufficiently small, so that the material behaves linearly and $h_1{}^b = 1$. This allows evaluation of $h_1{}^a$, and consequently, $h_2{}^a$. In subsequent two-step tests, strains will be chosen for ε^b which have already been evaluated in terms of h_1 and h_2, so that the value $h_1{}^b$ will be known. Through the above process, values can be obtained for h_∞, h_1, h_2, and a_ε at each of the temperatures of interest.

A process has been described which should be sufficient to allow for the development of a constitutive relationship of the form of Equation (19). The tests required to evaluate this relationship completely are given in Table 1. This series of tests must be conducted at a sufficient number of temperatures, probably three or four, to allow for adequate evaluation of the constitutive relationship over the full range of temperatures inherent in balloon flight.

Table 1. Proposed Test Matrix For Each Temperature

Test Number	One-Step Relaxation Test	Two-Step Relaxation Test	
	ε^a	ε^a	ε^b
1	ε_1*		
2	ε_2	ε_2	ε_1
3	ε_3	ε_3	ε_2
4	ε_4	ε_4	ε_3
5	ε_5	ε_5	ε_4

*ε_1 must be sufficiently small to allow only linear response.

V. DISCUSSION OF RESULTS

The viscoelastic nature of thin polyethylene film has been carefully detailed in the preceding sections of this report. Available data has been used to graphically demonstrate the nonlinear character of this material which has previously been modeled as either linearly elastic or viscoelastic. In addition, a test procedure has been described which will permit the evaluation of the various functions needed to produce an accurate nonlinear characterization.

In order to assess the importance of an accurate material characterization in the analysis of balloon stress and strain, a simplified comparison of the results obtained to date and previous results is revealing. Consider the problem of creep under constant stress conditions at -23°C. This would be somewhat analogous to the circumferential stress state of a balloon under float conditions. Since laboratory data is obtained in a relatively short period of time, then the corresponding material properties such as those reported by Alexander [9] would predict relatively small strains. If a stress of 6.89 MPa (1000 psi) is assumed then strain on the order of 1.5 percent would be predicted. However, a typical balloon must remain in this stressed state from 4 to 24 hours which will cause the strain to increase to as much as 2.8 percent, an increase of 86 percent from the original value. This estimate is based on the simplest linear viscoelastic model, Equation (42), which has already been shown to neglect the nonlinearities due to strain.

Another representative problem is that of relaxation under constant strain. This would be somewhat analogous to the meridional stress state of a balloon since the load tapes will tend to restrain deformations in this direction. If a strain of 1.5 percent is assumed, then a stress of 6.89 MPa (1000 psi) would be predicted from short term data. This stress would relax to 3.62 MPa (525 psi) after 24 hours which represents a decrease of 47 percent from the original value. In a balloon, this would cause a load redistribution and the tape force would have to increase to maintain meridional equilibrium.

Another problem of importance, especially during ascent, is the transient temperature history of the film. A method must be developed to take this event into account. This may account for the observations by several investigators that summer operations tend to have more apparent balloon failures than engineering flights conducted in the winter months. This may be caused by the substantially different strain distributions which would occur if a balloon is held in the launch configuration at different temperatures prior to ascent.

Each of these problems indicate that transient response of the film must be understood if the state of stress or strain is to be determined with sufficient accuracy to identify causes of balloon failure. In order to accomplish this, a selective set of laboratory tests directed by those responsible for the characterization is necessary to obtain realistic values of stress from strains measured in flight.

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